MENTHOL BEHAVIOR IN THE FILTER CIGARETTE: I. THE DEPENDENCE OF MENTHOL MIGRATION AND DELIVERY ON THE SITE OF APPLICATION. Lance J. Deutsch, Filter Products Division, Hoechst Celanese Corporation, P. O. Box 32414, Charlotte, North Carolina, 28232 USA.

SUMMARY

The behavior of menthol within a filtered cigarette was studied over time as a function of site of menthol application to the cigarette. Migration of menthol within the cigarette followed the expected pattern; menthol moved from areas of high concentration to low concentration. The plasticized filter continued to absorb menthol, which became largely unavailable to the smokestream, except for a small amount delivered via elution. Menthol deliveries were site-dependent intially but exhibited no significant differences after the menthol migration reached a steady state. Individual contributions of the filter and the tobacco column were determined and discovered to be independent of each other.

INTRODUCTION

Menthol is one of the most widely used flavorants in the tobacco industry. The volatility of menthol is a problem in terms of efficient application to the cigarette, retention during storage, and delivery upon smoking. Typical rates of utilization of delivered menthol relative to that applied to the cigarette range from 10 to 15%. Although these rates are low, the greater concern is variability over time, which can have a negative impact on smoker satisfaction.

This study was undertaken to determine both the diffusion of menthol within the cigarette and delivery of menthol in the mainstream smoke as a function of site of application to the cigarette and as a function of shelf life. This was accomplished by studying several sets of cigarettes prepared by shifting menthol application from tobacco columns to filter tip in 25% increments. The cigarettes were then allowed to age in sealed packs and sampled at intervals for smoking.

In a corollary to this study, mentholated filter tips were removed from their tobacco columns at controlled intervals, attached to non-mentholated tobacco columns, and smoked. Similarly, mentholated tobacco columns were detached from their filter tips and smoked on non-mentholated filter tips. In this way, the individual contributions to menthol delivery from the tobacco columns and the filter tips could be investigated.

EXPERIMENTAL

Six sets of cigarettes were manufactured to identical specifications (Table 1), with the exception of the percentage of menthol applied to the filter tips vs. the tobacco columns. Nominal menthol application was 3 mg per cigarette. A non-mentholated cigarette was also produced as a control and as a source of non-mentholated filter tips and tobacco columns. The cigarettes were non-ventilated. The filters were 21 mm in length and consisted of standard cellulose acetate tow bonded with 6% triacetin. The tobacco columns were 63 mm in length and contained a typical American tobacco blend.

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Menthol content of the filter tips and tobacco columns was determined immediately after manufacture and then periodically for the next eight months. At the same time, the cigarettes were smoked to determine menthol deliveries. These results were then correlated with the original site of menthol application as a function of time. The test schedule and scheme are given in Table 2. Testing was done more frequently during the early part of the study to provide more detailed information during the steepest part of the menthol transfer curve.

All cigarettes were smoked on a Filtrona SM350 machine according to Coresta protocols, with the exception that they were not conditioned. This was done in order to simulate actual smoking conditions as well as to minimize loss of menthol to evaporation. Twenty cigarettes were smoked through four Cambridge traps.

Water, nicotine, and menthol deliveries were determined via gas chromatographic analysis of alcoholic extracts of the smoked Cambridge filters. Water was quantified on a Poropak Q column and thermal conductivity detector. Nicotine and menthol were quantified on a 30 meter J&W DB-WAX megabore column (1.0μ film thickness) and flame ionization detector. A Hewlett Packard 5880A GC was used for all analyses.

Menthol in smoked and unsmoked (blank) filter tips was determined by GC analysis of alcoholic extracts of the filter tips. Menthol content of the tobacco columns was determined by duplicate aqueous distillations of the tobacco of two cigarettes, followed by GC analysis.

DISCUSSION

The migration of menthol within a cigarette is relatively predictable. Figure 1 shows the menthol content of the filter tip as a function of time for the samples (samples A and B are replicates of the 100% menthol on tobacco variant). As expected, the cigarettes to which menthol was applied mostly to the filter show a rapid loss from the filter, whereas the cigarettes to which menthol was mostly applied to the tobacco show a rapid increase in filter menthol. A slow but steady gain in filter menthol follows this initial equilibration period; this is due to the relatively greater solubility of menthol in plasticized cellulose acetate.

Mirroring this is the decrease in menthol from the tobacco columns (Figure 2). Initially, excess menthol from the filter tip rapidly transfers to the tobacco, creating a rise in menthol content for samples E and F. There follows a slow loss of menthol for all samples, as menthol is absorbed by the filter, maintaining a chemical potential between tobacco and filter. Note that sample D (menthol applied 50/50 filter/tobacco) exhibits very little menthol migration during the equilibration period.

The separation into two distinct groups is due to the difficulty of applying significant quantities of menthol to the filter. A material balance indicates that samples D, E, and F contained approximately 2.5 mg menthol, which is 0.5 mg short of nominal. Subsequent figures have been corrected for this shortage.

Menthol delivery is shown in Figure 3. Not surprisingly, the cigarettes with large amounts of menthol applied to the filter show a very high delivery for

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several days after manufacture. This is due to excess menthol at or near the surface of the filter fibers eluting into the smokestream. On the other hand, cigarettes A and B, where all menthol was applied to the tobacco, exhibit a slight increase in delivery during the first few days. A reasonable hypothesis for this behavior is that menthol initially in the tobacco that is lost to sidestream smoke migrates to the filter, where it is available to mainstream smoke via elution. In all cases, the menthol distributes between filter and tobacco in a very short time (<20 days) and menthol delivery for all cigarettes becomes nearly indistinguishable.

Note that sample C, to which 25% of the total menthol was applied to the filter, exhibits very little initial slope. This could have significance in terms of minimizing changes in menthol impact immediately after cigarette making.

Figure 4 describes the net gain or loss of filter menthol as a result of smoking. This corresponds to the deposition of menthol onto the filter via the particulate matter, less elution of both absorbed and deposited menthol from the filter during smoking. The equilibrium value is consistently around 0.2 mg of menthol.

Figure 5 demonstrates menthol utilization, which is defined as the percentage of menthol delivered relative to that applied during cigarette manufacture. As expected, utilization decreases over time due to irreversible absorption of menthol into the cellulose acetate filter.

The second part of this study examined the individual contributions of the filter tips and the tobacco columns to menthol behavior. At appropriate intervals, tips were removed from the mentholated cigarettes and smoked on non-mentholated columns (Case I). Similarly, the columns of the mentholated cigarettes were harvested and smoked on non-mentholated filter tips (Case II).

This "mix and match" approach resulted in several interesting observations. Figure 6 shows the menthol delivery from the Case I samples. After equilibration, all cigarettes deliver approximately 0.2 mg menthol, which is only available via elution from the filter. There is a downward trend over time, which is in direct contrast to the upward trend in tip menthol over time (Figure 1). Diffusion of menthol into the bulk polymer of the fibers over time evidently renders the dissolved menthol inaccessible to the smokestream. The decrease in menthol transfer due to depleted tobacco menthol results in a decrease in the excess surface menthol on the fiber, and a decrease in delivery.

Conversely, menthol delivery from the Case II samples exhibits a steady state sometime after day 76 (Figure 7). Menthol deposition onto the non-mentholated filter is also relatively constant after this date. This is understandable in view of the fact that, unlike elution of menthol from the filter, distillation of menthol from the tobacco is complete.

It is interesting to note that if the independent deliveries for a given filter tip/tobacco column are combined (Figure 8), the plots obtained are indistinguishable from those of the original cigarettes (Figure 3). It can be concluded that either: 1) there is no interaction between filter tip menthol content and menthol delivery from tobacco smoke, or; 2) the two are exactly offsetting due to a complex interaction between menthol elution and particulate matter deposition. The latter seems unlikely, although additional experiments are necessary to rule it out altogether.

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CONCLUSIONS

1. During the first 6 to 8 weeks, tobacco column menthol content is dependent upon the original site of application and will increase or decrease accordingly. It then slowly (but steadily) decreases. Filter tip menthol content behaves in a similar but opposite fashion.

2. Within certain limits, the stabilized menthol content of the filter is independent of the original site of application.

3. During the stabilization period (first 2 to 3 weeks), menthol delivery is dependent upon application site. After stabilization, all cigarettes deliver approximately equivalent amounts of menthol, which slowly decreases with time.

4. During the stabilization period, applying some menthol to the filter (between 25% and 50%) minimizes changes in menthol delivery with time.

5. After stabilization, the net gain in the menthol content of the filter tips after smoking is small (<.3mg) and constant.

6. After stabilization, menthol utilization decreases over time at a relatively constant rate and is independent of the original application site.

7. A significant amount of menthol elutes from the filter when smoke passes through the filter. This is more than offset by the quantity of menthol in the particulate matter trapped by the filter.

8. Menthol delivery due to the tobacco smoke is apparently independent of the menthol content of the filter tip.

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TABLE 1

CIGARETTE PROPERTIES

Length	84 mm
Tip Length	21 mm
Tip Weight	170 mg
Tip Circumference	25.0 mm
Tip Pressure Drop	68 mm H_2O
Ventilation	None
Tobacco	Standard Domestic Blend
Plasticizer	6% (Nominal) (Triacetin)
Menthol (Target)	3 mg/cig
Deliveries	Full Flavor

TABLE 2

SAMPLE IDENTIFICATION

	TIP MENTHOL % of TOTAL	COLUMN MENTHOL % of TOTAL
A	0	100
B	0	100
C	25	75
D	50	50
E	75	25
F	100	0
G	0	0

SAMPLES TESTED

Primary - A, B, C, D, E, F, G
CASE I (Filter/Tobacco Column) - BG, CG, DG, EG, FG
CASE II (Filter/Tobacco Column) - GB, GC, GD, GE, GF

TESTING SCHEDULE

DAYS - 0, 2, 6, 13, 20, 34, 48, 76, 104, 160, 233

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Figure 1

TIP MENTHOL

UNSMOKED CIGARETTE

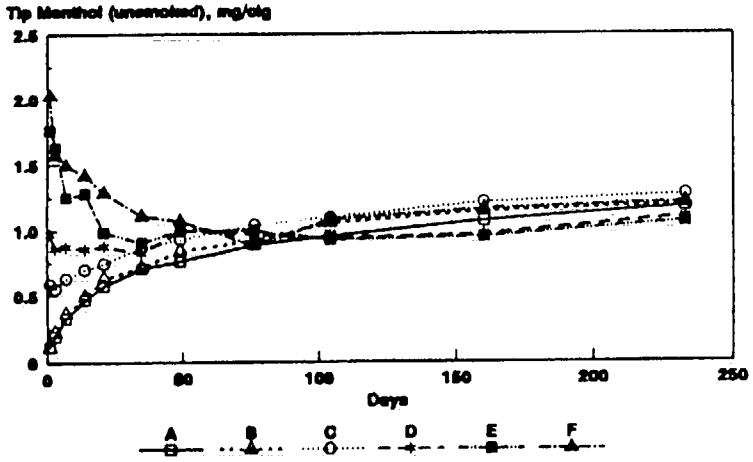

Figure 2

COLUMN MENTHOL

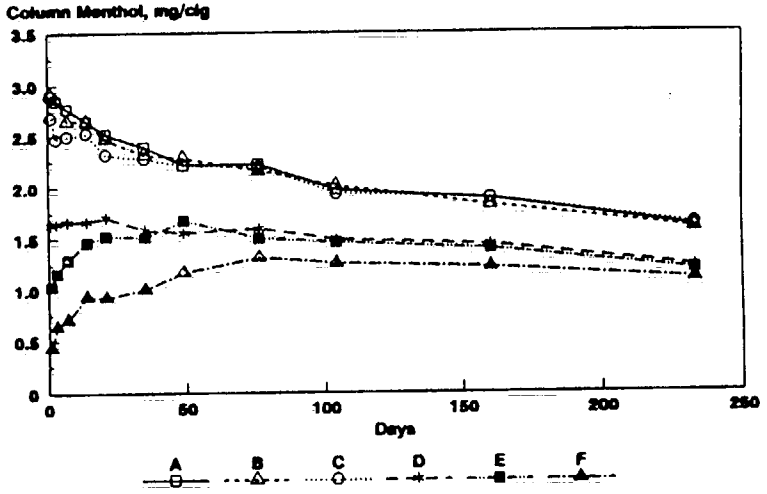

Figure 3

MENTHOL DELIVERY, CORRECTED

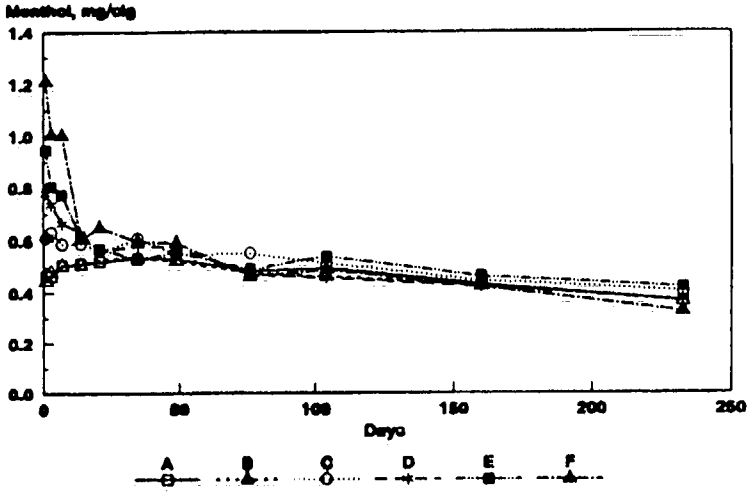

Figure 4

FILTER NET MENTHOL GAIN OR LOSS:

PRIMARY CIGARETTES

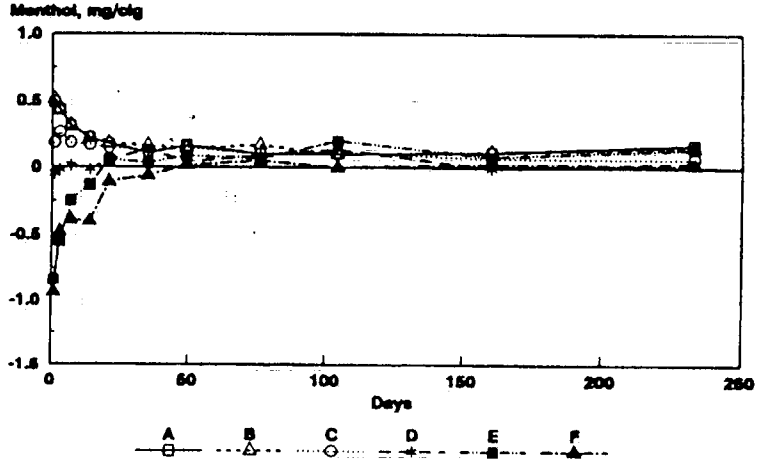

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Figure 5
Menthol Utilization:
Percent Delivery of Applied

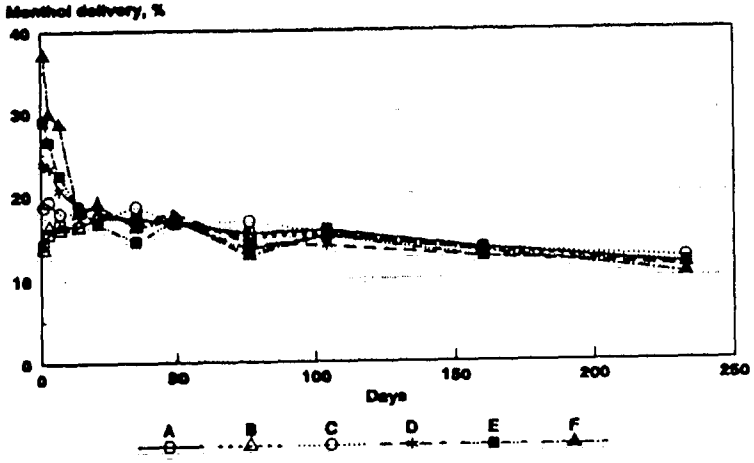


Figure 6
MENTHOL DELIVERY

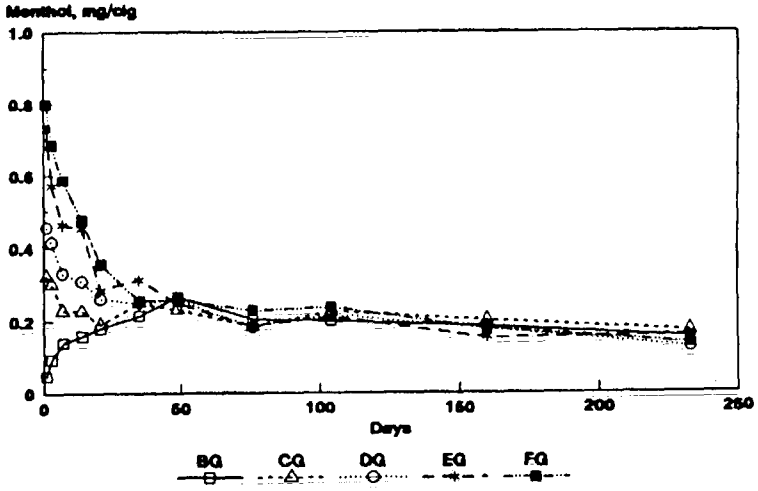


Figure 7
MENTHOL DELIVERY

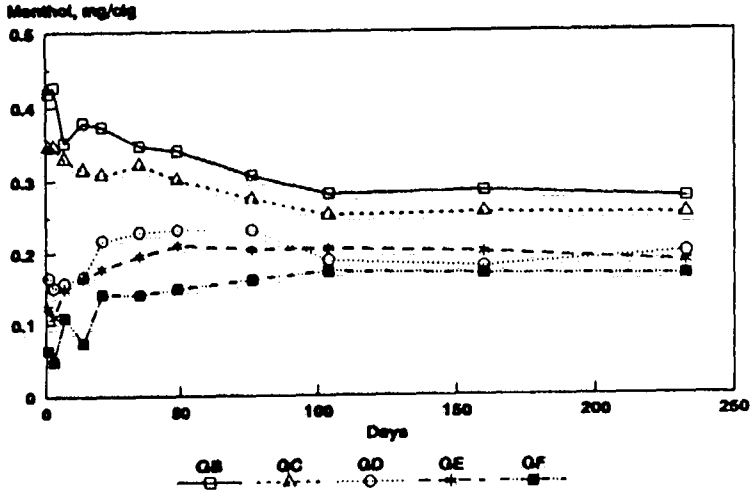
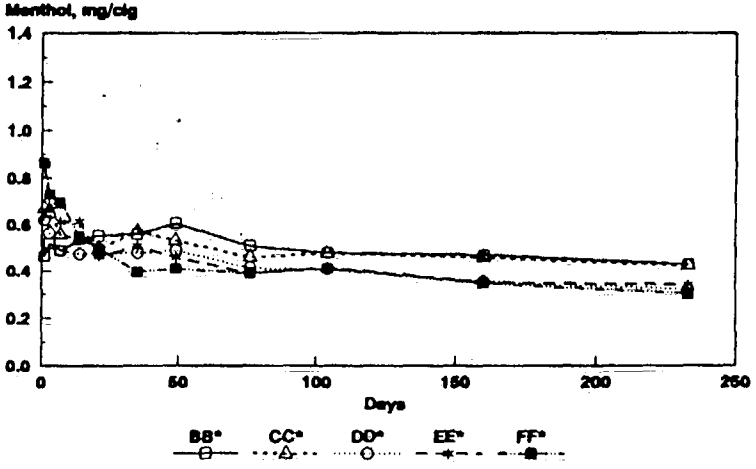


Figure 8
RECONSTITUTED MENTHOL DELIVERY
TIP DELIVERY PLUS COLUMN DELIVERY



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